Final Report for the ARPA/DSO Contract No: N00014-93-1-0664

Contract Title: Single Crystals for High Average Power Solid State Lasers

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Project Summary:

In this contract we have achieved the design and construction of a three zone horizontal Bridgman gradient freeze furnace capable to produce large Nd:YAG single crystal slabs for high average power diode pumped lasers. We have acquired and constructed all the major components of the furnace and the system is under assembling. In addition, we have investigated nonlinear optical borate crystals with high optical damage threshold. This includes RNB (RbNbB2O6), BCBF (BaCaBO3F). We also developed a highly efficient Nd laser crystal, SFAP (Sr5(PO4)3F) for microchip lasers for both 1.06 and 1.32 μm as well as intracavity doubling at 529 and 664 nm applications.

I. Introduction:

The development of solid state laser has reached to a critical point that the basic theory is well understood and the hardware design has also reached its maturity. As a consequence, any further improvement of laser performance rely almost entirely on the material properties of the active lasing medium. One of them is the optical coating technology. The tight control to meet coating specification as well as to produce high optical damage resistant coating are essential to optimize the pump cavity. The other critical issue is the laser crystal itself. In order to achieve the desired lasing output and efficiency, the quality of the gain medium must be at its best. For Nd laser hosts, it is necessary to maximize the product of emission cross-section and minimizing loss due to scattering or absorption. It is well known fact that using the same pumping chamber, the laser output can vary by more than a factor of two depending entirely on the laser crystal quality. This is particularly critical in high average power operations. The laser can either fail to deliver the power as needed or be catastrophysically destroyed by high power pumping. In this contract, we started to tackle the material issues in three aspects:

(1) Design a new crystal growth furnace to produce large size Nd:YAG slabs capable to produce high power via either conventional or diode pumping. Despite Nd:YAG is the most developed laser crystal, large size slab is still unavailable at the present time due to technical limitations.

(2) Search for new laser hosts whixh can provide distinct improvement over Nd:YAG for specific lasing arrangement.

(3) Search for new nonlinear optical crystals with sufficient nonlinear coefficient and high damage threshold capable to handle high power frequency conversions. Most of all, it is desirable to find congruent melting compounds so that large crystals can be grown with reduced cost.

II. Design New Crystal Growth Furnaces:

As mentioned in the introduction that despite the maturity of the conventional Czochralski melt pulling technique, it is not capable to produce sufficient size crystal boules for the needed slabs for high average power application. Under this contract, we have designed an unique three zone horizontal Bridgman gradient freeze furnace to produce such crystal. The furnace is designed to produce 30 x 15 x 1 cm3 Nd:YAG slab. The basic furnace frame work has been build. It include four major components:

(1) Furnace frame: Since it is more expensive to construct vacuum sealed system with delicate moving mechanism, we decided to use nitrogen gas purging to maintain inert growth chamber which is needed to protect the growth crucible. The body frame consists an open chamber with water cooling system to remove the heat. A specially designed three sapphire rod horizontal translation system is used to carry the weight of the crucible and charge through the gradient freeze furnace. The rod is placed underneath at the cooler part of furnace and can take up to 1000°C without significantly loss its strength. The translation is driven by two separated motors with high and low speed. The low speed motor can move the charge from 0.1 to 100 mm/hr which is adequate for the growth.

(2) Power supply: There was a lot of consideration to select the workable heating design. The conventional graphite or tungsten heating design is relatively cheap at beginning but more expensive in real operation. The heating rate is slow and difficult to control sharp temperature gradient. The induction heating is much more expensive at beginning but it is easier to maintain and fast in response. We decided to use the induction heating for this system. In order to provide the necessary temperature gradient, we use three Le Pel 35 KW induction generators with three specially designed induction coils to provide the necessary power.

(3) Crucible and insulation: This is the most critical and also the most difficult part of the entire furnace design. The difficulty lies on the fact that YAG melts near 2000°C and there are few materials can handle such melt. Traditional Ir metal container is out of the question due to the excess cost since the YAG crystal sticks to the crucible well and the container can only use once and will be destroyed in order to remove the crystal. We decided to use a thin wall molybdenum crucible and it can only weakly couple to the induction power. The main portion of the power is deliver to a 1/4" thick tungsten plate suscepter which in turn heats the crucible. After the crucible is determined, the next step is the insulation. The material should be insulating, refractory, high strength, light weight, none reactive and none cracking. As expected, the most difficult one is to find materials that can 2000°C temperature cycling without cracking. Since there is no material with such property, we decided to use a unique composite insulation design. The inner part is made of BN which has very low thermal expansion coefficient whereas the outer part is made of porous Zirconia.

(4) Power control: A unique cascade temperature control system will be used to control the growth temperature. The temperature of this three zone gradient freeze furnace will be measured by a tungsten-rhenium thermocouple at each zone. The control feed back will then be cascaded into the power feed-back loop for the final control. The advantage of this design is high resolution. It also provide both high and low power limits to protect the furnace from thermocouple failure.

During this contract we have acquired all the major hardewares of the furnace. These components of the furnace has installed in the Laboratory for assembly. Power and water cooling system have connected. We expect to complete the assembly before the end of this year and the furnace will under several tests for power supply, temperature stability and the integraty of the insulation before actual growth. Details of these work are described in our new proposal. The basic layout of the equipment is illustrated in the following pictures.

III. Search for New Laser Hosts:

Even though Nd:YAG has the most universal application in all aspects of the solid state lasers, it is by no means the best material for every specific applications. This is especially true now a days with great advances of semiconductor laser diodes as pump source. To achieve high lasing efficiency in diode pumped Nd lasers requires the Nd host to have high product of emission cross-section and fluorescence lifetime. In addition, long fluorescence lifetime is also desirable for Q-switching operation. High absorption cross section at the diode pumping wavelengths (near 810 nm) will also help to reduce sample thickness which will make cooling more efficient.

Because of these reasons, there is a renew interest on Nd:YVO_4 and its isomorph, $GdVO_4$. Unfortunately, both crystals are difficult to grow and they all have perfect cleavage planes making fabrication delicate. We initiated the research on Nd doped fluoro-apatite structure compounds, namely, $Ca_5(PO_4)_3F$ (FAP), $Sr_5(PO_4)_3F$ (SFAP) and $Sr_5(VO_4)_3F$ (SVAP). All three compounds melt near congruently and can be grown by standard Czochralski pulling technique in large high quality single crystals. Although the general spectroscopic and lasing properties among the three are similar, there is a distinct superiority of SFAP over the other two due to longer fluorescence lifetime (300 μs), shorter emission wavelength (1.0585 μm) and cleaner spectroscopic features.

Direct comparison of lasing properties at 1.06 μm region shows that Nd:SFAP is at least comparable if not superior over Nd:YVO_4 and Nd:$GdVO_4$. Moreover, the unique large Stark splittings at both $^4F_{3/2}$ and $^4I_{11/2}$ levels provide essential single transition. The same large Stark splitting also occurs at $^4I_{13/2}$ level, so that the emission at 1.328 μm is also single transition. Most important of all is that the branching ratio for $^4I_{13/2}$ level for fluoro-apatite compounds is the highest among all known materials. The emission cross-section of SFAP for 1.0585 and 1.328 μm are 5.4 and 2.3 x 10^{-19} cm^2, respectively. Efficient lasing (≈ 50% slope efficiency) has achieved at 1.328 μm.

Efficient lasing in two wavelengths in SFAP provides unique opportunities in laser design. This is the first time that a single diode pumped laser can do ALL the following:

(1) Efficient lasing at both 1.0585 and 1.328 μm (for cable TV transmitter application).
(2) Efficient frequency doubling (with KTP and CTA) to 529 and 664 nm.
(3) Simultaneous lasing at both 1.0585 and 1.328 μm.
(4) SFM of the two to generate 589 nm (Na light) from a single laser source
(5) OPO pumped by 1.0585 and 1.328 μm using KTP and CTA, respectively, to cover the entire spectrum range from 1.5 to 5.0 μm without gaps.

This is the first time that multi-purpose uses can be achieved with one single laser system. No other material can do it at present time. This is quite a revolution in diode pump solid state laser technology with great potentials for military, medical, industrial and even consumer applications.

At present, we have the state-of-the-art technology on SFAP growth. The crystal has both smoke and slip dislocation problems. We are able to remove smoke using a unique post growth annealing procedure and eliminate the slip dislocations by readjust growth orientation. We are quite confident that Nd:SFAP will be used in many applications.

IV. Search for New Nonlinear Optical Crystals:

Because all the useful radiative energy transitions for both transitional metal and rare earth ions are located in the near IR region, to extend it to either shorter (UV and visible) and longer

(mid- to far-IR) wavelength region, it is necessary to use nonlinear frequency conversion scheme. Nonlinear optical crystals are required for such conversion process. Despite near three decades of research, only a handful of nonlinear crystals are useful and all of them have some shortcomings depending on the application. For many of the more recently developed crystals such as KTP, BBO and LBO, the main problem is the slowness of the growth process.

Under this contract, we also initiated the search for new nonlinear optical materials. Our goal is to seek materials with high non-linearity, high transparency and non-hygroscopic. To achieve phase matching condition, it is necessary to have materials with adequate birefrigence and low dispersion. Although nitrate and carbonate have distinct advantages in terms of birefrigence, they have serious problems of hygroscopy for the nitrates and growth difficulty for the carbonates. Since borate, in general, does not have nearly the same problems, they have been the choice for new nonlinear material search. Both BBO and LBO are examples of such success.

We also look into the borate compounds for potential nonlinear harmonic conversions. Here are two examples of our effort. The first one is RNB (RbNbB2O6) and the second one is BCBF (BaCaBO3F).

RNB is one of the compound with the general formula of XYB2O6 where X = K, Rb, Cs and Y = Nb, Ta. It is orthorhombic with mm2 in symmetry. The compound melts incongruently and has to be grown by flux method. It has the unique Nb-B conjugate ring structure which provide the in-plane (a-b plane) optical nonlinearity. One of the unique feature of this compound is large delocalization of Nb along the c-axis direction which, in principle, should provide large optical nonlinearity. Unfortunately, the Nb atoms are in zigzag arrangement so that the large nonlinearity along c-axis is canceling to each other. We have made an effort to ordering the Nb arrangement by using equal amount of alkali ions with distinct difference in ionic radius such as Na, Rb pairs or K, Cs pairs without success. The Nb containing conjugate ring structure also reduces the birefrigence so that KNB is not phase matchable at 1.06 µm. Both RNB and CNB can only have type I phase matching. The in plane nonlinearity is only slightly higher than BBO. The transparency cut off is at 350 nm, but they are not phase matchable below 1 µm. The crystal is nonhygroscopic but has two distinct sets of cleavage planes making device fabrication somewhat difficult. Our feeling of these compounds at present time is that although RNB and its isomorphs have interesting and unique structure, their nonlinear performace does not show distinct superiority over existing materials such as KTP, BBO and LBO. There is also no distinct advantages on growth and device fabrication. It is a useful material but will be difficult to compete in commercial market.

To be useful in commercial application, one of the key factor is the cost. At present, all the available nonlinear crystals except for LiNbO3 are grown by flux method. The growth rate is slow and the yield is low. It is highly desirable to find a congruent melting nonlinear compound so that the cost of growth can be greatly reduced. We can even trade off the optical nonlinearity with crystal length. Following this idea, we initiate the growth of BCBF (BaCaBO3F) single crystals. Despite the compositional complexity, the material melts congruently around 950°C. We have successfully produced this crystal by conventional Czochralski pulling technique. The compound is uniaxial and trigonal in symmetry. The optical nonlinearity is comparable to LBO and the transparency cut-off is 270 nm. Both the nonlinear coefficient and the phase matching condition are under investigation. The compound has a plannar BO3 structure to ensure adequate birefrigence. It is likely that the material is phase matchable for the 670 nm red diode to the UV (335nm). Although the crystal is growable by Czochralski pulling, the actual growth is not as easy as expected. We found by surprise that the melt is somewhat reactive to the Ir-crucible. The crystal is light brown color and the melt shows serious decomposition. Colorless BCBF crystal can only be produced from Pt crucibles. Because of the BO3 planar structure, the crystal has a weak (0001) cleavage plan. The preferred growth direction is also along c-axis. We found that the crystal is sensitive to thermal shock and easy to crack. For reasons unknown, it is not easy to

use weight control for growth. We are continuing the investigation. The crystal does offer many attractive features making the investigation worthwhile.

V. Conclusion:

In this contract we have conducted the investigations in three specific areas with the aim to search and grow bettter laser and nonlinear optical crystals for high average power applications. Our primary goal is to design and construct a unique three zone horizontal Bridgman gradient freeze furnace capable to produce large Nd:YAG single crystal slabs which can not be produced by conventional growth techniques. So far we have acquired and constructed all the necessary major components of the furnace and the system is under assembling. We expect that the system will be operational by the end of this year. In addition, we have investigated two nonlinear optical borate crystals RNB ($RbNbB2O6$) and BCBF ($BaCaBO3F$). RNB is phase matchable to 1.06 µm Nd laser with adequate damage threshold. The nonlinearity is comparable to BBO and much less than KTP. The two sets of cleavage planes will make the crystal fabrication difficult. BCBF is a new congruent melting nonlinear borate compound. Even though the nonlinearity is low comparable to that of LBO, it can phase match to shorter wavelengths due to higher birefrigence. It has potential to be the material for frequency doubling of the 670 nm red diode to the UV region. During this contract, we also developed a highly efficient Nd laser crystal, SFAP ($Sr5(PO4)3F$). Although the primary interest of this crystal is for diode pumped microchip lasers, we fond that the crystal also works well as laser rods in conventional flash lamp pumped cavities. The same rods can also be pumped with laser diodes. We have initiated the design of a laser diode side pumped system. Our preliminary result shows that the material can compete favorably with all the existing diode pumped laser hosts including Nd:YAG, Nd:YLF, Nd:YVO4, Nd:GdVO4 and the recently developed Nd:LSB crystal ($LaSc3(BO3)4$) at the 1.06 µm and 530 nm region. Since Nd:SFAP also works exceptionally well at 1.32 µm and 664 nm region, it has great potential as the material of choice for medium to low power diode pumped miniature lasers.

VI. Appendix: Phtographs of the Proposed Horizontal Bridgman Growth Furnace:

Fig. 1: Overall View of the Furnace System:

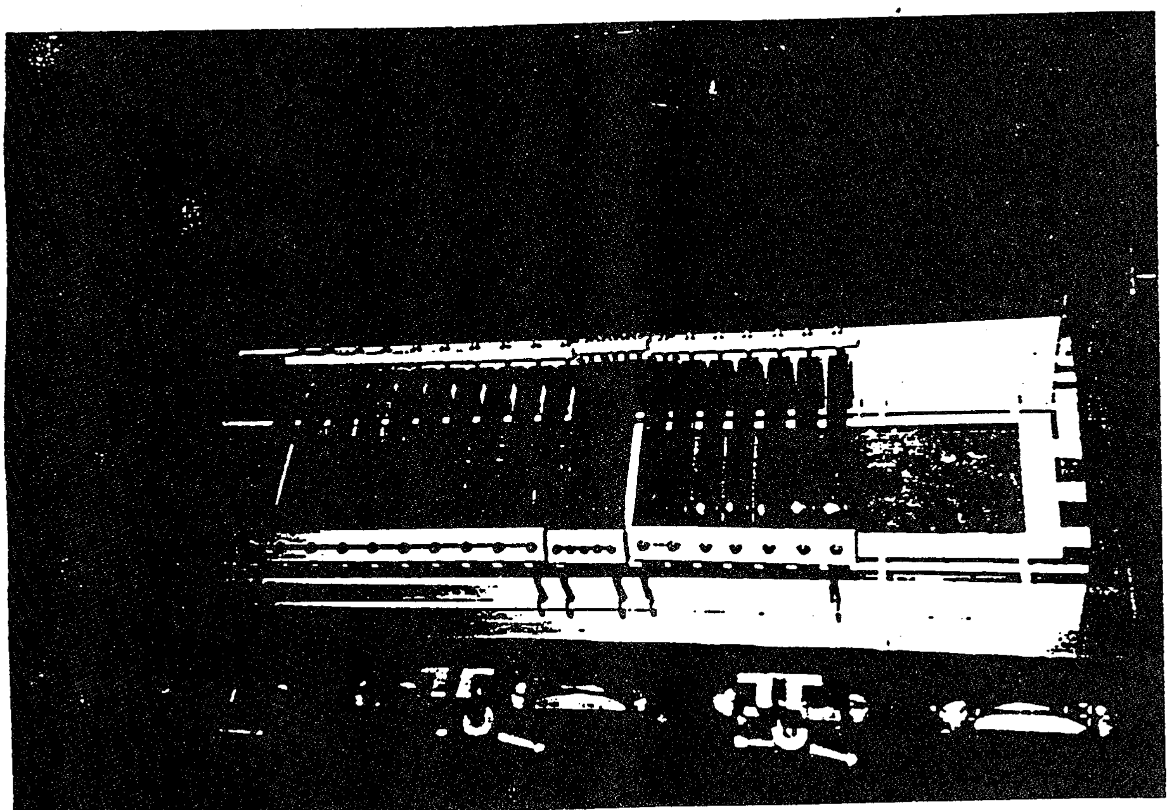

Fig. 2: Detail view of the three zone induction coils and tungsten plate: (insulation removed)

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